

**SYSTEM AND METHOD FOR FORMING A MEMBRANE
ELECTRODE ASSEMBLY FOR FUEL CELLS**

BACKGROUND OF THE INVENTION

-- This application is a National Stage of PCT KR2005/001410 filed on May 13, 2005 which claims priority to Korean Application No. 10-2004-0033791 filed on May 13, 2004 and Korean Application No. 10-2005-0039942 filed May 13, 2005, all of which are incorporated by reference, as if fully set forth herein.--

(a) Field of the Invention

The present invention relates to a system for manufacturing a membrane electrode assembly (MEA) for a fuel cell.

(b) Description of the Related Art

A fuel cell is a device for generating electric power using electrons generated during an oxidation-reduction reaction of oxygen and hydrogen. A unit cell of the fuel cell is formed by coating an anode and a cathode on both sides of an electrolyte membrane that is made of a high polymer substance and is generally called a membrane electrode assembly. Hydrogen or methanol, as a fuel, is supplied to the anode, and it reacts to generate hydrogen ions, and the hydrogen, passing the high polymer electrolyte membrane, reacts with oxygen in the cathode, thereby generating pure water.

1 Such reactions occur in the membrane electrode assembly, and the
2 membrane electrode assembly is manufactured by coating layers of anode
3 catalyst material (typically, Pt or Pt/Ru) and cathode catalyst material
4 (typically, Pt) on both sides of a high polymer membrane.

5 One known method for manufacturing the membrane electrode
6 assembly is a method in which a paste is formed by mixing a catalyst material,
7 a proton conductive binder material, and a water or alcohol group solvent, the
8 formed paste is coated on a carbon cloth or a carbon paper, and the carbon
9 cloth or paper coated by the paste is applied to the proton conductive
10 electrolyte membrane through a heat welding. This method is a kind of
11 indirect coating method, and it has a problem in that since the catalyst
12 material is not distributed with a uniform thickness on a surface of a porous
13 carbon cloth or a porous carbon paper when the catalyst material is coated
14 thereon, but permeates into the porous carbon cloth or a porous carbon paper,
15 a percentage of use of the catalyst while operating the membrane electrode
16 assembly is decreased, and thereby an overall performance is deteriorated.
17 In addition, since the already formed electrode layer is secondly heat-welded
18 onto the proton conductive layer, a manufacturing process becomes
19 complicated, and an interfacial surface of the electrolyte material and the
20 catalyst layer is discontinuously formed.

21 Furthermore, another method for manufacturing the membrane
22 electrode assembly is a method in which an electrode paste is formed by
23 mixing a catalyst material, a proton conductive binder material, and a water or
24 alcohol group solvent, and then coating this paste onto a surface of the proton

1 conductive electrolyte membrane through a direct transfer coating method,
2 such as a screen printing method. Although this method, imitating a general
3 printing manufacturing process, is advantageous to patterning and is suitable
4 for mass production, a swelling phenomenon may occur where a volume of
5 the high polymer electrolyte membrane increases in all directions when the
6 water or alcohol group solvent comes into contact with the high polymer
7 electrolyte membrane, so that it becomes difficult to obtain a uniform coated
8 layer, a loss of catalyst solution increases, and it becomes difficult to regulate
9 a viscosity of the solution.

10 As another method for manufacturing the membrane electrode
11 assembly, there is a wet-type method in which a slurry solution is formed by
12 mixing a catalyst material, a proton conductive binder material, and a large
13 quantity of a water or alcohol group solvent, and a catalyst layer is formed by
14 spraying this slurry solution onto a gas diffusion layer or a high polymer
15 electrolyte membrane using a spray device. Also, there is a dry-type method
16 in which a catalyst material and a proton conductive binder material are mixed
17 with each other without using a solvent, and a catalyst layer is formed by
18 injecting the mixture of the catalyst material and the proton conductive binder
19 material using an electrostatic attraction force. These methods have an
20 advantage that a continuity of an interfacial surface and a percentage of use
21 of the catalyst can be improved since a thin catalyst layer is formed on the
22 gas diffusion layer or the high polymer electrolyte membrane through a direct
23 coating method.

24 However, in the wet-type method, although the catalyst material and

1 the proton conductive material can be uniformly mixed since a large quantity
2 of solvent is used, there is a drawback that it is difficult to obtain a uniform
3 coated layer due to a swelling of a membrane when a coating is performed
4 directly onto the membrane. In addition, in the dry-type method, there is a
5 problem in that it is difficult to perform a coating using the electrostatic
6 attraction force since the membrane is an insulator and it is difficult to
7 uniformly mix catalyst particles and the proton conductive binder material.

8 Therefore, a new system for manufacturing the membrane electrode
9 assembly is needed, in which a uniform catalyst layer may be formed by
10 uniformly spraying the catalyst solution onto the high polymer layer to improve
11 a performance of a fuel cell and to allow mass production of the membrane.

12 **SUMMARY OF THE INVENTION**

13 The present invention has been provided to solve the above-
14 mentioned problems, and it is an object of the present invention to provide a
15 system and a method for manufacturing a membrane electrode assembly for
16 a fuel cell that can increase uniformity of a catalyst layer and minimize a
17 swelling phenomenon of a high polymer electrolyte membrane by adopting
18 devices for preheating an carrying gas and a catalyst solution.

19 A system for manufacturing a membrane electrode assembly for a fuel
20 cell according to an embodiment of the present invention includes a catalyst
21 solution preheating device, an carrying gas preheater, a cathode catalyst
22 solution spray nozzle, and an anode catalyst solution spray nozzle. The
23 catalyst solution preheating device preheats a cathode catalyst solution and
24 an anode catalyst solution. The carrying gas preheater preheats an carrying

1 gas. The cathode catalyst solution spray nozzle is supplied with the cathode
2 catalyst solution preheated by the catalyst solution preheating device and the
3 carrying gas preheated by the carrying gas preheater, and is configured to
4 spray the supplied cathode catalyst solution. The anode catalyst solution
5 spray nozzle is supplied with the anode catalyst solution preheated by the
6 catalyst solution preheating device and the carrying gas preheated by the
7 carrying gas preheater, and is configured to spray the supplied anode catalyst
8 solution.

9 The catalyst solution preheating device may heat the cathode catalyst
10 solution and the anode catalyst solution at a temperature in a range of $0.6 \times BP$
11 to $0.95 \times BP$, where BP is a boiling point of the solvent of the catalyst solution.

12 The catalyst solution preheating device may include a cathode
13 catalyst solution preheater preheating the cathode catalyst solution and an
14 anode catalyst solution preheater preheating the anode catalyst solution.

15 The cathode catalyst solution preheater may heat the cathode catalyst
16 solution at a temperature in a range of $0.6 \times BP$ to $0.95 \times BP$, where BP is a
17 boiling point of the solvent of the catalyst solution.

18 The anode catalyst solution preheater may heat the anode catalyst
19 solution at a temperature in a range of $0.6 \times BP$ to $0.95 \times BP$, where BP is a
20 boiling point of the solvent of the catalyst solution.

21 The cathode and anode catalyst solution spray nozzles may operate
22 to alternately spray the catalyst solution.

23 The carrying gas preheater may heat the carrying gas at a
24 temperature higher than boiling points of the cathode catalyst solution and the

1 anode catalyst solution.

2 The carrying gas may be one of argon, helium, nitrogen, and air.

3 A method for manufacturing a membrane electrode assembly for a
4 fuel cell according to an embodiment of the present invention includes
5 preheating the cathode catalyst solution, preheating the anode catalyst
6 solution, preheating the carrying gas, spraying the preheated cathode catalyst
7 solution through a cathode catalyst solution spray nozzle using the preheated
8 carrying gas, and spraying the preheated anode catalyst solution through an
9 anode catalyst solution spray nozzle using the preheated carrying gas.

10 The spraying of the preheated cathode catalyst solution and the
11 spraying of the preheated anode catalyst solution may be alternately
12 performed.

13 In the preheating cathode catalyst solution, the cathode catalyst
14 solution may be heated at a temperature in a range of $0.6 \times BP$ to $0.95 \times BP$,
15 where BP is a boiling point of the solvent of the catalyst solution.

16 In the preheating anode catalyst solution, the anode catalyst solution
17 may be heated at a temperature in a range of $0.6 \times BP$ to $0.95 \times BP$, where BP is
18 a boiling point of the solvent of the catalyst solution.

19 In the preheating of the carrying gas, the carrying gas may be heated
20 at a temperature higher than the boiling points of the cathode catalyst solution
21 and the anode catalyst solution.

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2 **BRIEF DESCRIPTION OF THE DRAWINGS**

3 FIG. 1 is a schematic diagram of a system for manufacturing a
4 membrane electrode assembly for a fuel cell according to an embodiment of
5 the present invention.

6 FIGs. 2 and 3 comparatively show voltage and power characteristics
7 of a membrane electrode assembly manufactured by a system for
8 manufacturing a membrane electrode assembly for a fuel cell according to an
9 embodiment of the present invention and by a conventional system.

10 **DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS**

11 Hereinafter, embodiments of the present invention will be explained
12 with reference to the drawings.

13 Referring to FIG. 1, a system for manufacturing a membrane
14 electrode assembly for a fuel cell according to an embodiment of the present
15 invention includes a catalyst solution preheating device 10 preheating a
16 cathode catalyst solution and an anode catalyst solution.

17 The catalyst solution preheating device 10 is supplied with the
18 cathode catalyst solution and the anode catalyst solution respectively from a
19 cathode catalyst solution storage container 11 and an anode catalyst solution
20 storage container 15, preheats the supplied solutions, and discharges the
21 preheated solutions.

22 In the present embodiment, the catalyst solution preheating device 10
23 includes a cathode catalyst solution preheater 13, preheating the cathode
24 catalyst solution, and an anode catalyst solution preheater 17, preheating the

1 anode catalyst solution. However, the catalyst solution preheating device 10
2 can be achieved as a single preheater that is configured to separately preheat
3 the cathode catalyst solution and the anode catalyst solution.

4 The cathode catalyst solution preheater 13 is supplied with the
5 cathode catalyst solution from the cathode catalyst solution storage container
6 11 and preheats the supplied cathode catalyst solution. The cathode catalyst
7 solution is a solution of a cathode catalyst material that is formed by mixing
8 the cathode catalyst material with a solvent or by dispersing the cathode
9 catalyst material into a solvent, and is used to form a cathode of a membrane
10 electrode assembly of a fuel cell. For example, the cathode catalyst solution
11 can be a known cathode catalyst solution.

12 The anode catalyst solution preheater 17 is supplied with the anode
13 catalyst solution from the anode catalyst solution storage container 15 and
14 preheats the supplied anode catalyst solution. The anode catalyst solution is
15 a solution of an anode catalyst material that is formed by mixing the anode
16 catalyst material with a solvent or by dispersing the anode catalyst material
17 into a solvent, and is used to form an anode of a membrane electrode
18 assembly of a fuel cell. For example, the anode catalyst solution can be a
19 known anode catalyst solution.

20 The cathode and anode catalyst solution preheaters 13 and 17 can be
21 realized by a known preheater.

22 The cathode and anode catalyst solution preheaters 13 and 17 may
23 respectively preheat the cathode catalyst solution and the anode catalyst
24 solution at a temperature lower than the boiling points of the solvents of the

1 cathode catalyst solution and the anode catalyst solution. Solvents used for
2 the catalyst solutions may be water or alcohol group solvents, and the alcohol
3 group solvents may be methanol, ethanol, n-propanol, isopropyl alcohol, 1-
4 buthanol, 2-buthanol, isobutyl alcohol, or the like. The cathode and anode
5 catalyst solution preheaters 13 and 17 may preheat the cathode catalyst
6 solution and the anode catalyst solution at a temperature in a range of 60% to
7 95% of the boiling point of the solvent of the catalyst solution, i.e., in a range
8 of $0.6 \times \text{BP}$ to $0.95 \times \text{BP}$, where BP is a boiling point of the solvent of the catalyst
9 solution. If the catalyst solution is preheated at a temperature lower than
10 $0.6 \times \text{BP}$, a sufficient preheating effect cannot be obtained, and if the catalyst
11 solution is preheated at a temperature higher than $0.95 \times \text{BP}$, vaporization of
12 the solvent may occur so that the catalyst material may block an outlet
13 passageway of a spray nozzle.

14 The cathode catalyst solution preheated by the cathode catalyst
15 solution preheater 13 is supplied to a cathode catalyst solution spray nozzle
16 19, and the anode catalyst solution preheated by the anode catalyst solution
17 preheater 17 is supplied to an anode catalyst solution spray nozzle 21.

18 An carrying gas preheater 33 is supplied with an carrying gas from an
19 carrying gas storage container 31 and preheats the supplied carrying gas.
20 The carrying gas is a gas for carrying the cathode and anode catalyst solution
21 to spray nozzles 19 and 21, and for example, it may be argon (Ar), helium
22 (He), nitrogen (N₂), or air.

23 The carrying gas preheater 33 may preheat the carrying gas at a
24 temperature higher than the boiling points of the solvents of the catalyst

1 solutions such that the carrying gas can evaporate the solvent of the catalyst
2 solution simultaneously with the spraying of the catalyst solution. For example,
3 the carrying gas preheater 33 may preheat the carrying gas at a temperature
4 higher than or equal to a temperature corresponding to 110% of the boiling
5 point (BP) of the solvent of the catalyst solution, i.e., $1.1 \times \text{BP}$. Since the
6 carrying gas is preheated at a temperature higher than the boiling point of the
7 solvent of the catalyst solution by the carrying gas preheater 33, the catalyst
8 material may be sprayed onto a high polymer electrolyte membrane 1 after
9 the solvent of the catalyst solution is evaporated by the heat of the carrying
10 gas.

11 The carrying gas preheated by the carrying gas preheater 33 is
12 supplied to the cathode catalyst solution spray nozzle 19 and the anode
13 catalyst solution spray nozzle 21.

14 For example, the cathode catalyst solution preheater 13, the anode
15 catalyst solution preheater 17, and the carrying gas preheater 31 may be
16 realized as a known preheater.

17 The cathode catalyst solution spray nozzle 19 is supplied with the
18 cathode catalyst solution preheated by the cathode catalyst solution
19 preheater 13 and the carrying gas preheated by the carrying gas preheater 31,
20 and is configured to spray the supplied cathode catalyst solution.

21 The anode catalyst solution spray nozzle 21 is supplied with the
22 anode catalyst solution preheated by the anode catalyst solution preheater 17
23 and the carrying gas preheated by the carrying gas preheater 31, and is
24 configured to spray the supplied anode catalyst solution.

1 The cathode catalyst solution sprayed by the cathode catalyst solution
2 spray nozzle 19 and the anode catalyst solution sprayed by the anode
3 catalyst solution spray nozzle 21 are coated respectively on each side of the
4 high polymer electrolyte membrane 1 that is disposed between pattern frames
5 3. Accordingly, the membrane electrode assembly in which the cathode and
6 the anode are formed on each side of the electrolyte membrane is formed.

7 According to an embodiment of the present invention, since the
8 cathode catalyst solution, the anode catalyst solution, and the carrying gas
9 are supplied to the spray nozzles after having been respectively preheated,
10 and the preheated solution is then sprayed, a catalyst layer can be more
11 uniformly formed and a swelling phenomenon in the electrolyte membrane
12 can be minimized.

13 FIGs. 2 and 3 comparatively show voltage (V) and power (Watt)
14 characteristics of a membrane electrode assembly manufactured by a system
15 for manufacturing a membrane electrode assembly for a fuel cell according to
16 an embodiment of the present invention and by a conventional system.

17 In FIGs. 2 and 3, graphs connecting “●” and “▲” illustrate relations
18 between a current density A/cm^2 and a power density $watt/cm^2$, and graphs
19 connecting “○” and “△” illustrate relations between the current density A/cm^2
20 and a cell voltage V. Also, in FIG. 2, “●” and “○” relate to data of a membrane
21 electrode assembly of CCM (catalyst coated membrane) type manufactured
22 by a system according to an embodiment of the present invention, and “▲”
23 and “△” relate to data of a membrane electrode assembly of CCM type
24 manufactured by a conventional system. Moreover, in FIG. 3, “●” and “○”

1 relate to data of a membrane electrode assembly of a type in which a catalyst
2 is coated on a gas diffusion layer (GDL) manufactured by a system according
3 to an embodiment of the present invention, and “▲” and “△ ” relate to data of
4 a membrane electrode assembly of a type in which a catalyst is coated on a
5 GDL manufactured by a conventional system.

6 As is already known in the art, it is advantageous that an amount of
7 decrease of the voltage and the power density depending on an increase of a
8 current density is small. Referring to FIGs. 2 and 3, an amount of decrease of
9 the voltage and the power density depending on an increase of a current
10 density in the membrane electrode assembly manufactured by a system
11 according to an embodiment of the present invention is smaller than in the
12 membrane electrode assembly manufactured by a conventional system, and
13 in particular, in a relatively high current density region, a difference of
14 amounts of decrease of the voltage and the power density is relatively great.
15 Accordingly, the membrane electrode assembly manufactured by a system
16 and a method according to embodiments of the present invention, i.e., the
17 membrane electrode assembly manufactured by spraying the preheated
18 catalyst solution, has favorable voltage and power density characteristics, and
19 in particular, has especially favorable voltage and power density
20 characteristics in a high current density region, when compared to the
21 membrane electrode assembly manufactured by a conventional system.

22 In more detail, since at least a portion of the solvent of the catalyst
23 solution can be removed during the spraying because the preheating process
24 is performed, catalyst material, having been dried to some extent, is coated

1 on a surface of the high polymer electrolyte membrane or a gas diffusion
2 layer. Accordingly, shrinkage of the high polymer electrolyte membrane or the
3 gas diffusion layer due to the solvent can be decreased, so that the catalyst
4 solution can be coated more uniformly. Furthermore, with the decrease in the
5 amount of solvent, the phenomenon that the catalyst particles are closely
6 banded together by the solvent is substantially decreased. In addition, if the
7 dried catalyst particles are coated on the high polymer electrolyte membrane
8 or the gas diffusion layer, a sufficient porosity among catalyst particles may
9 exist so that material transfer characteristics can be improved. Still
10 furthermore, since the dried catalyst material is sprayed on the gas diffusion
11 layer, it can be minimized that the catalyst material is permeated into the gas
12 diffusion layer together with the solvent so that the usability of the catalyst is
13 increased and good voltage and power characteristics can be obtained.

14 The cathode catalyst solution spray nozzle 19 is installed to a
15 transportation device 25, and the transportation device 25 transports the
16 cathode catalyst solution spray nozzle 19 in at least one direction so that the
17 cathode catalyst solution spray nozzle 19 can equally spray the cathode
18 catalyst solution onto the high polymer electrolyte membrane 1. For such an
19 operation of the transportation device 25, the transportation device 25 may be
20 installed to a frame 23.

21 Similarly, the anode catalyst solution spray nozzle 21 is installed to a
22 transportation device 29, and the transportation device 29 transports the
23 anode catalyst solution spray nozzle 21 in at least one direction so that the
24 anode catalyst solution spray nozzle 21 can equally spray the anode catalyst

1 solution onto the high polymer electrolyte membrane 1. For such an operation
2 of the transportation device 29, the transportation device 29 may be installed
3 to a frame 27.

4 Operations of the transportation devices 25 and 29 can be controlled
5 by a pre-inputted program, and a control unit (not shown) for storing and
6 executing the pre-inputted program can be provided within the transportation
7 devices 25 and 29 or can be provided outside the transportation devices 25
8 and 29.

9 In the embodiment of the present invention, the cathode and anode
10 catalyst solution spray nozzles 19 and 21 operate to alternately spray the
11 catalyst solution. Accordingly, while one of the cathode and anode catalyst
12 solution spray nozzles 19 and 21 is spraying the catalyst solution, the other
13 spray nozzle is dried by the preheated carrying gas. Therefore, the catalyst
14 solution can be more effectively sprayed and the sprayed catalyst solution
15 may form a uniform layer.

16 Such operations of the cathode and anode catalyst solution spray
17 nozzles 19 and 21 can be controlled by a pre-inputted program, and a control
18 unit for storing and executing the pre-inputted program can be provided within
19 the cathode and anode catalyst solution spray nozzles 19 and 21 or can be
20 provided outside the cathode and anode catalyst solution spray nozzles 19
21 and 21.

22 The control unit may include a microprocessor, a memory, and other
23 necessary hardware and software components as will be understood by
24 persons skilled in the art, to permit the control unit to execute the control

1 function as described herein.

2 In addition, the control unit for controlling the transportation devices 25
3 and 29 and the control for controlling the cathode and anode catalyst solution
4 spray nozzles 19 and 21 can be realized as a single unit.

5 Meanwhile, a method for manufacturing a membrane electrode
6 assembly for a fuel cell according to an embodiment of the present invention
7 can be performed by a system for manufacturing a membrane electrode
8 assembly for a fuel cell according to an embodiment of the present invention.

9 The method for manufacturing the membrane electrode assembly
10 according to an embodiment of the present invention includes preheating a
11 cathode catalyst solution, preheating an anode catalyst solution, preheating
12 an carrying gas, spraying the preheated cathode catalyst solution through a
13 cathode catalyst solution spray nozzle using the preheated carrying gas, and
14 spraying the preheated anode catalyst solution through an anode catalyst
15 solution spray nozzle using the preheated carrying gas.

16 The spraying of the preheated cathode catalyst solution and the
17 spraying of the preheated anode catalyst solution are alternately performed.

18 While the present invention has been described in connection with the
19 most practical exemplary embodiments, it is to be understood that the
20 invention is not limited to the disclosed embodiments, but, on the contrary, is
21 intended to cover various modifications and equivalent arrangements
22 included within the spirit and scope of the appended claims.

23 Throughout this specification and the claims which follow, unless
24 explicitly described to the contrary, the word "comprise", or variations such as

1 "comprises" or "comprising", will be understood to imply the inclusion of stated
2 elements but not the exclusion of any other elements.

3 According to embodiments of the present invention, since the cathode
4 catalyst solution, the anode catalyst solution, and the carrying gas are
5 supplied to the spray nozzles after they are preheated, the catalyst layer can
6 be formed more uniformly and a swelling phenomenon of the high polymer
7 electrolyte membrane can be minimized. Moreover, since the spraying of the
8 preheated cathode catalyst solution and the spraying of the preheated anode
9 catalyst solution are alternately performed, the catalyst layer can be formed
10 more uniformly.

11